

# Effects of the Magnetic Field on the Relaxation of Small Particle Systems

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## Abstract

We study the effect of a magnetic field on the thermal relaxation of non-interacting small monodomain particle systems particles with a distribution of anisotropy constants and random easy-axes directions. Numerical calculations of the relaxation curves for different distribution widths, and under different magnetic fields  $H$  and temperatures  $T$ , have been performed in the framework of a two-state approximation. We show how the obtained data can be analyzed in terms of an modified  $T\ln(t/\tau_0)$  scaling from which the field dependence of the mean relaxing energy barriers can be extracted, a microscopic information which is not easily obtainable by other methods.

*Key words:* Classical spin models, Numerical simulation studies, Fine-particle systems, Magnetic aftereffects.

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## 1 Introduction

The progressive miniaturization of the elementary recording units for magnetic recording has put forward the problem of stability of magnetization against thermal activation. [1,2] Therefore, the study of the thermal relaxation of the magnetization in fine particle systems is of crucial interest to ascertain the stability of the recorded information. Although the first studies in this field date back to the 40's [3–5], at present, there is no complete model to account for all the experimental findings. One of the points that has not been completely clarified is the influence of a magnetic field in the relaxation.

We want to account for the experimental studies on the relaxation of small-particle systems, which essentially measure the acquisition of magnetization of an initially demagnetized sample under the application of a magnetic field.

[6–10] In this kind of experiments, the field modifies the energy barriers of the system that are responsible for the time variation of the magnetization, as well as the final state of equilibrium towards which the system relaxes. Here, we will demonstrate that the previously proposed  $T \ln(t/\tau_0)$  scaling of the relaxation data at zero field [6,11–16] can also be extended to systems relaxing in an applied magnetic field. We will show how information about the microscopic energy barriers responsible for the relaxation can be obtained from this procedure.

The paper is organized as follows. In Sec. 2, we present the basic features of model showing how the distribution of energy barriers of the system is influenced by the application of a magnetic field with the help of the concept of effective energy barrier distribution. In Sec. 3, we derive the equation governing the time dependence of the magnetization from a master rate equation for a Two-State Approximation (TSA). The results of numerical calculations based on the above mentioned equation are presented in Sec.4. There, we present the  $T \ln(t/\tau_0)$  scaling of relaxation curves at a given magnetic field, discussing its range of validity. We also study the possibility of a scaling at different fields and fixed temperature, and its applications. Finally in Sec. 5 we resume the main conclusions of the article.

## 2 Model

We consider an ensemble of randomly oriented, noninteracting single-domain ferromagnetic particles of volume  $V$  and magnetic moment  $\mathbf{M} = M_s V \mathbf{m}$  with uniaxial anisotropy. To take into account the spread of particle volumes in real samples, we will assume that the particles anisotropy constants  $K$  are distributed according to a logarithmic-normal distribution

$$f(K) = \frac{1}{\sqrt{2\pi}K\sigma} e^{-\ln^2(K/K_0)/2\sigma^2} , \quad (1)$$

The energy of a particle is determined by the orientation of  $\mathbf{M}$  with respect to the external magnetic field  $\mathbf{H}$  and to the easy-axis direction  $\mathbf{n}$ . Using the angular coordinates defined in Fig. 1, it can be written as

$$\bar{E} = \frac{E}{VK} = -\cos^2(\theta) - 2h \cos(\theta - \psi) . \quad (2)$$

where we have defined the reduced field  $h \equiv H/H_c$  and  $H_c = 2K/M_s$  as the critical field for an aligned particle. We have concentrated on the two dimensional case ( $\mathbf{M}$  lying in the plane formed by  $\mathbf{H}$  and  $\mathbf{n}$ ) since the energy maxima and minima can be calculated analytically only in this case. In Fig.

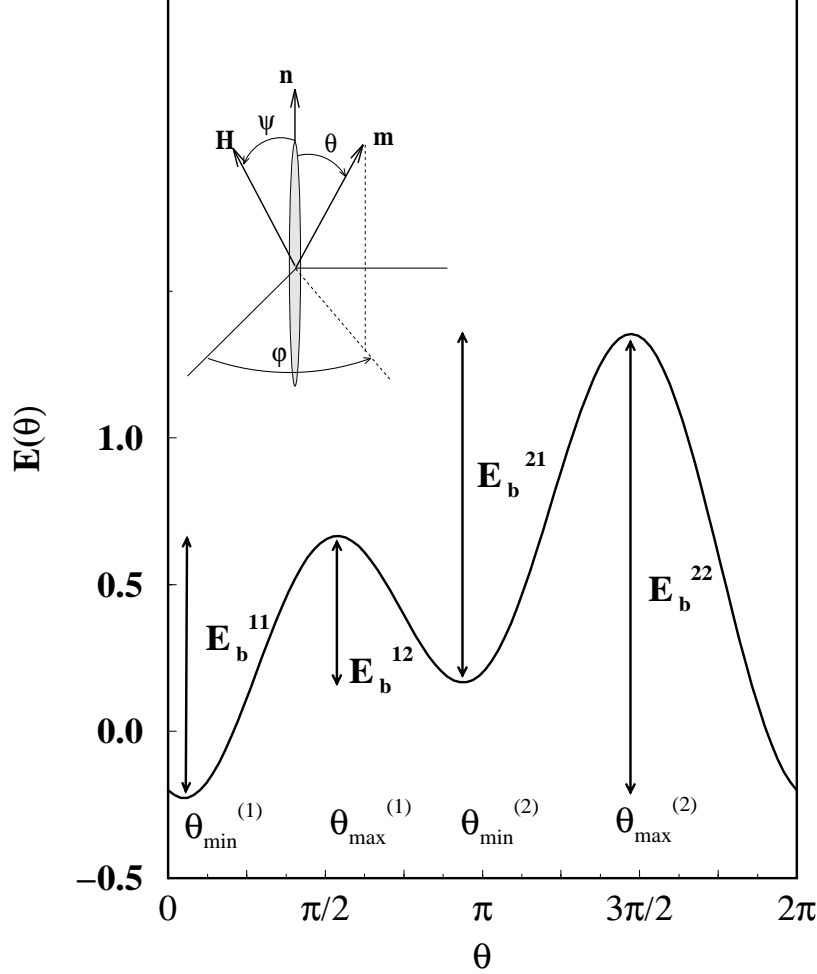


Fig. 1. Energy function  $E(\theta, \psi)$  as a function of the angle between the magnetization vector  $\mathbf{m}$  and the magnetic field  $\mathbf{h}$ , for  $\mathbf{m}$  in the plane formed by the easy-axis and the magnetic field ( $\varphi = 0$ ), as given by Eq. (2). The plot is for a particle whose easy-axis  $\mathbf{n}$  forms an angle  $\psi = 30^\circ$  with  $\mathbf{h}$ , and  $H = 0.3$ . We have used the following notation to designate the extrema of the energy:  $\theta_{\min}^1$  and  $\theta_{\max}^1$  refer to the extrema closer to the field direction while  $\theta_{\min}^2$  and  $\theta_{\max}^2$  refer to those farther from the direction of the field. The four possible energy barriers between them are  $E_b^{ij} \equiv E(\theta_{\max}^i) - E(\theta_{\min}^j)$ . Inset: Schematic representation of the quantities involved in the definition of the system. The easy-axis of the particles  $\mathbf{n}$  are in the x-z plane forming an angle  $\psi$  with the magnetic field  $\mathbf{H}$ , which points along the z axis.  $\theta$  and  $\varphi$  are the spherical angles of the magnetization vector  $\mathbf{M}$ .

1, we show the variation of the energy with  $\theta$  for a typical case, defining in the same figure the notation for the energy barriers and extrema.

The magnetic field modifies the energy barriers of the system depending on the particle orientation and anisotropy value, and, consequently, changes the original energy barrier distribution [17,18]. In order to understand the qualitative change of  $f_{\text{eff}}$  with  $h$ , we have numerically calculated  $f_{\text{eff}}(E_b)$  for a system of randomly oriented particles with  $f(K)$ , for different widths  $\sigma$  and  $K_0 = 1$ , and several values of the magnetic field  $h$ . The calculation has been performed by making energy barriers histograms for a collection of 10 000 particles. The results are given in Fig. 2. In all the cases, we observe the progressive splitting of the original distribution  $f(E_b^0)$  in two subdistributions of high and low barriers as  $h$  increases from zero. The field tends to make deeper one of the minima, therefore increasing the two energy barriers for rotation of  $\mathbf{M}$  out of the field direction, while the other two are reduced. In this way, the global effect of  $h$  is a splitting of  $f(E_b)$  towards lower and higher values of  $E_b$ . As  $h$  attains the critical value  $h_c$  for the particles with smaller  $K$ , a peak of zero or almost zero energy barriers starts to appear (see for example the curves for  $H = 0.5$  in the case  $\sigma = 0.5$ ); while most of the non-zero barriers are distributed according to a distribution identical to  $f(E_b^0)$ , but centered at higher energies. The higher the width of the distribution  $\sigma$ , the lower the  $h$  at which the lowest energy barriers start to be destroyed by the field. The random orientation of the anisotropy axes smears out the distributions and makes the minima less pronounced than for a system with aligned particles due to the spread in particle orientations, this effect being more noticeable at high  $h$ . In Sec. 4, we will discuss how these results affect the time dependence

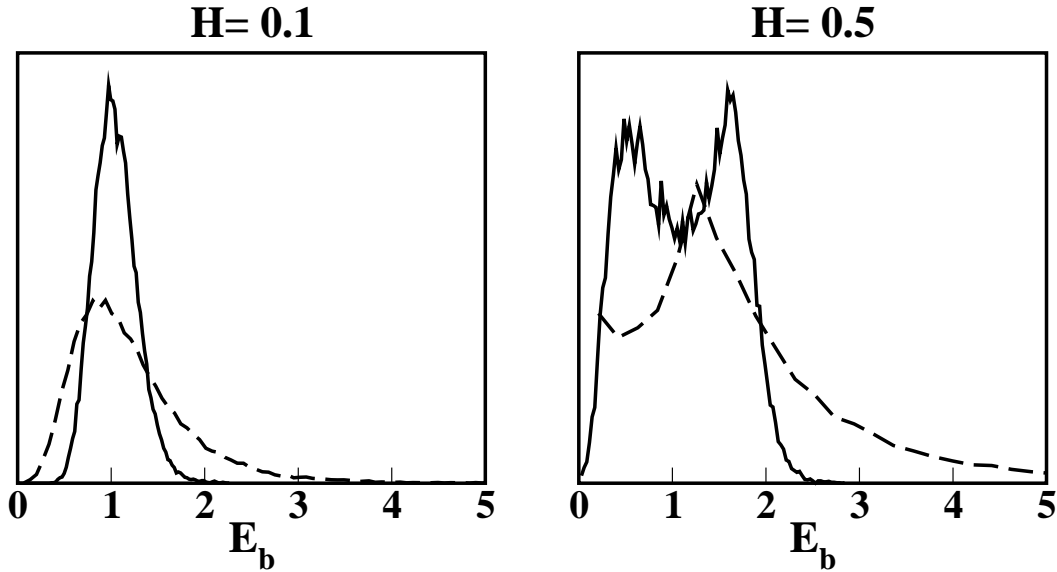


Fig. 2. Effective energy barrier distributions for particles with random orientations of anisotropy axes and a lognormal distribution of anisotropy constants of  $\sigma = 0.2$  (continuous lines),  $\sigma = 0.5$  (dashed lines), for values of  $H$  as indicated in the figures.

of magnetization in relaxation experiments.

### 3 Relaxation curves in the presence of a magnetic field

In order to derive an equation for the time dependence of the magnetization we propose a simple dynamical model that can be solved analytically in terms of intrinsic parameters. Since usually, in experiments performed in real samples the thermal energy  $k_B T$  is smaller than the anisotropy energy  $KV$ , we will concentrate on the high energy barrier (low  $T$ ) regime. In this case, it is justified to consider only transitions between the two energy minima of each particle, and to truncate the continuum of possible states for the magnetization to the two energy minima [19,20](TSA). The master equation for  $P_i$ , the probability to find the magnetization of the particle in the minimum  $i$ , can be written as[21]

$$\frac{dP_i}{dt} = \sum_{k=1,2} \sum_{j \neq i} \{w_{ji}^{(k)} P_j - w_{ij}^{(k)} P_i\} , \quad (3)$$

where  $w_{ij}^{(k)}$  stands for the transition rate for a jump from the state  $i$  to the state  $j$  separated by the maximum  $k$  (see Fig. 1). Our choice for these quantities takes into account the energy barrier separating the initial minimum  $i$  from the final state  $j$

$$w_{ij}^{(k)} = \frac{1}{\tau_{ij}^{(k)}} = \frac{1}{\tau_0} e^{-E_b^{ki} \beta} , \quad (4)$$

where  $\tau_0^{-1}$  is the attempt frequency and  $\beta = 1/k_B T$ .

Taking into account the normalization condition  $P_1 + P_2 = 1$ , one can easily solve Eq. (3) for  $P_1$  and  $P_2$  as a function of time

$$\begin{aligned} P_1(t) &= \frac{1 - e^{\beta \varepsilon} e^{-t/\tau}}{1 + e^{\beta \varepsilon}} \\ P_2(t) &= \frac{e^{\beta \varepsilon} (1 + e^{-t/\tau})}{1 + e^{\beta \varepsilon}} . \end{aligned} \quad (5)$$

The time-dependence of the system is thus characterized by an exponential function with a single relaxation time  $\tau$  that takes into account all possible probability fluxes

$$\tau^{-1} = \tau_0^{-1} \left( e^{-\beta E_b^{22}} + e^{-\beta E_b^{12}} \right) \left( 1 + e^{\beta \varepsilon} \right) , \quad (6)$$

where  $\varepsilon = E_{\min}^1 - E_{\min}^2$  is a measure of the asymmetry of the energy function.

The time dependence of the magnetization of the particle is then finally given by:

$$\begin{aligned} m(t; K, \psi) &= \cos[\theta_{\min}^1(\psi)]P_1(t) + \cos[\theta_{\min}^2(\psi)]P_2(t) \\ &= \bar{m}_{TS}(K, \psi) + [m_0 - \bar{m}_{TS}(K, \psi)]e^{-t/\tau(K, \psi)}. \end{aligned} \quad (7)$$

In this equation,  $\bar{m}_{TS}(K, \psi)$  is the equilibrium magnetization in the TSA, and  $m_0$  is the initial magnetization. If we have an ensemble of randomly oriented particles and a distribution of anisotropy constants  $f(K)$ , then the relaxation law of the magnetization is given by

$$m(t) = \int_0^\infty dK f(K) \int_0^\pi d\psi m(t; K, \psi). \quad (8)$$

This will be the starting point for all the subsequent numerical calculations of the relaxation curves and magnetic viscosity.

## 4 Numerical calculations

### 4.1 Relaxation curves: $T \ln(t/\tau_0)$ scaling and normalization factors

In this section, we present the results of numerical calculations of the magnetization decay based on Eq. (8). For the sake of simplicity, we have assumed zero initial magnetization  $m_0 = 0$ , so particles have initially their magnetic moments at random and evolve towards the equilibrium state  $m_{\text{eq}}$ . In the following, we will use dimensionless reduced variables for temperature and time, defined as  $T/T_0$  and  $t/\tau_0$ , with  $T_0 = E_0/k_B$  and  $E_0$  the value of the energy at which  $f(K)$  is centered. In Fig. 3, we show the results of the numerical calculations for a system with  $\sigma = 0.5$  and  $H = 0.5$  and temperatures ranging from 0.02 to 0.2. In the left hand panel, we present the original relaxations normalized to the equilibrium magnetization value that has been computed in the TSA. Normalization is essential in order to compare relaxations at different temperatures [14], especially at low fields where the temperature dependence of the equilibrium magnetization is more pronounced.

Our next goal is to investigate the possibility of scaling relaxation curves at different  $T$  in a given magnetic field with the scaling variable  $T \ln(t/\tau_0)$ , in the spirit of our previous works [11–14]. Let us briefly recall the basic points of

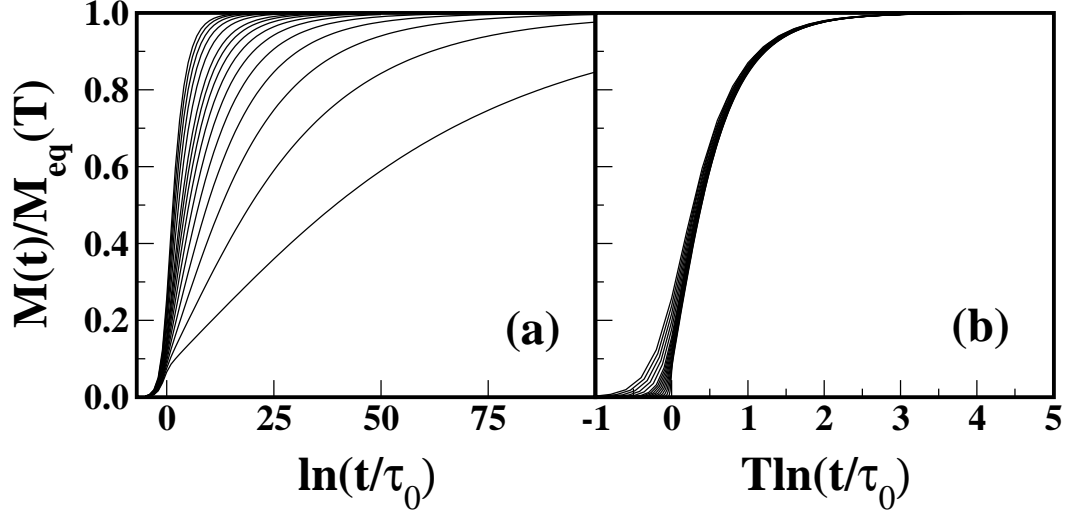


Fig. 3. (a) Relaxation curves for an ensemble of particles with randomly oriented anisotropy axes and the distribution  $f(K)$  of Eq. 1 of width  $\sigma = 0.5$  and  $K_0 = 1$  calculated by numerical integration of Eq. (8). Reduced temperatures  $T/T_0$ , starting from the lowermost curve, range from 0.01 to 0.1 with 0.01 increments, and from 0.1 to 0.2 with 0.02 increments. The applied field is  $H = 0.5$ . The original relaxation curves have been normalized to the equilibrium magnetization  $m_{TS}(T)$ . (b) Relaxation curves of panel (a) plotted as a function of the scaling variable  $T \ln(t/\tau_0)$  to obtain the master relaxation curve.

the method. The time dependence of the remanent magnetization of a system with a distribution of energy barriers  $f(E)$  can be written as

$$M(t) = M_0 \int_0^\infty dE f(E) e^{-t/\tau(E)} \simeq M_0 \int_{E_c(t)}^\infty dE f(E), \quad (9)$$

with a characteristic relaxation time given by the Arrhenius-Neél theory as  $\tau(E) = \tau_0 \exp^{E/T}$ . Then, the relaxation law for the magnetization will be a function of the parameter  $E_c(t) = T \ln(t/\tau_0)$  if the Boltzmann exponential factor in (9) can be approximated by a step function centered at  $E_c(t)$ . Then, as was shown in [11], the requirement for the validity of this approximation is that the characteristic width of  $f(E)$  is of the order of the width of the step-like function  $e^{-t/\tau(E)}$ , which can be calculated as  $\lambda = eT$  ( $e$  is the Napier number). This implies low enough  $T$  or energy distributions spread over a wide range of energies (large  $\sigma$ ).

In Fig. 3b, we show the original relaxation curves of Fig. 3a as a function of the scaling variable  $T \ln(t/\tau_0)$ . The collapse of the curves into a single master curve indicates that scaling is accomplished also in the presence of a magnetic field. Only at short times, the higher  $T$  curves depart from the master curve because in this  $T$  range the scaling requirements are no longer accomplished. According to the requirements for  $T \ln(t/\tau_0)$  scaling presented

in the preceding paragraph, in the presence of a magnetic field, the validity of the scaling hypothesis at a given  $T$  depends on the width of the effective energy barrier distribution (see Fig. 2). From simulations of master curves obtained at different  $H$  (not presented in Fig. 3), we observe that, the higher the field, the better the scaling of the curves is in the long time region and the worse at short times. This observation holds independently of the value of  $\sigma$ , indicating that it is a consequence of the application of a magnetic field. This can be understood with the help of the effective energy barrier distribution introduced in Sec. 2. As was shown in Fig. 2,  $H$  widens  $f_{\text{eff}}(E)$  and shifts the lowest energy barriers towards the origin, giving rise to a subdistribution of almost zero energy barriers that narrows with increasing  $H$ , and, consequently, the requirements for  $T \ln(t/\tau_0)$  scaling are worse fulfilled at small  $T \ln(t/\tau_0)$  values. On the contrary, as it was shown in section 2,  $H$  broadens the high energy tail of energy barriers that contribute to the relaxation,  $f(E_b^{22})$ , improving the scaling requirements at large  $T \ln(t/\tau_0)$  values.

#### 4.2 Scaling of relaxation curves at different magnetic fields

Another interesting point is the possibility of finding an appropriate scaling variable to scale relaxation curves at different fields for a given  $T$ , in a way similar to the case of a fixed field and different temperatures, in which  $T \ln(t/\tau_0)$  is the appropriate scaling variable. In a first attempt, we will study the effect of  $h$  on a system with random anisotropy axes and distribution of anisotropies  $f(K)$  of width  $\sigma$ , Eq. (1). Low temperature relaxation rates, obtained by performing the logarithmic time derivative of  $M(t)$

$$S(t) = \frac{dM(t)}{d[\ln(t)]} \simeq T f(E_c(t, T)) , \quad (10)$$

which is the coefficient of magnetic viscosity  $S(t)$  are presented in Fig. 4, for  $\sigma = 0.2, 0.5$ . From this expression, we see that when  $S$  is evaluated by taking the derivative of the master relaxation curve with respect to  $T \ln(t/\tau_0)$ , the resulting curve is proportional to the effective energy barrier distribution. First, let us notice that the qualitative shape of the viscosity curves is not distorted by  $H$ . The magnetic field simply shifts the position of the maxima towards lower values of  $T \ln(t/\tau_0)$  and narrows the width of the peaks, these effects being similar for both studied  $\sigma$ . The energy corresponding to the maximum relaxation rate ( $E_b^{\text{max}}$ ) decreases with increasing  $h$ , following the decrease of the smallest energy barriers (see Fig. 5), with an almost linear dependence on  $h$ . As can be clearly seen in Fig. 5, the dependence of  $E_{\text{max}}$  on  $h$  (in symbols) follows that of the lowest energy barriers for particles oriented at  $\psi = \pi/4$  and with  $K = K_{\text{max}}$  (dashed lines), which have been evaluated numerically from Eq. 2. Let us also notice that  $S_{\text{max}}$  becomes almost constant



below  $h_0$  (the value at which the field starts to destroy the lowest energy barriers) and low enough  $T$  (see the position of the maxima in the curves of Fig. 4). indicates that the relaxation curves at different  $h$  and fixed  $T$  may be brought to a single curve by shifting them along the  $T \ln(t/\tau_0)$  axis in accordance to the  $E_{\max}$  variation. The resulting curves are displayed in Fig. 6

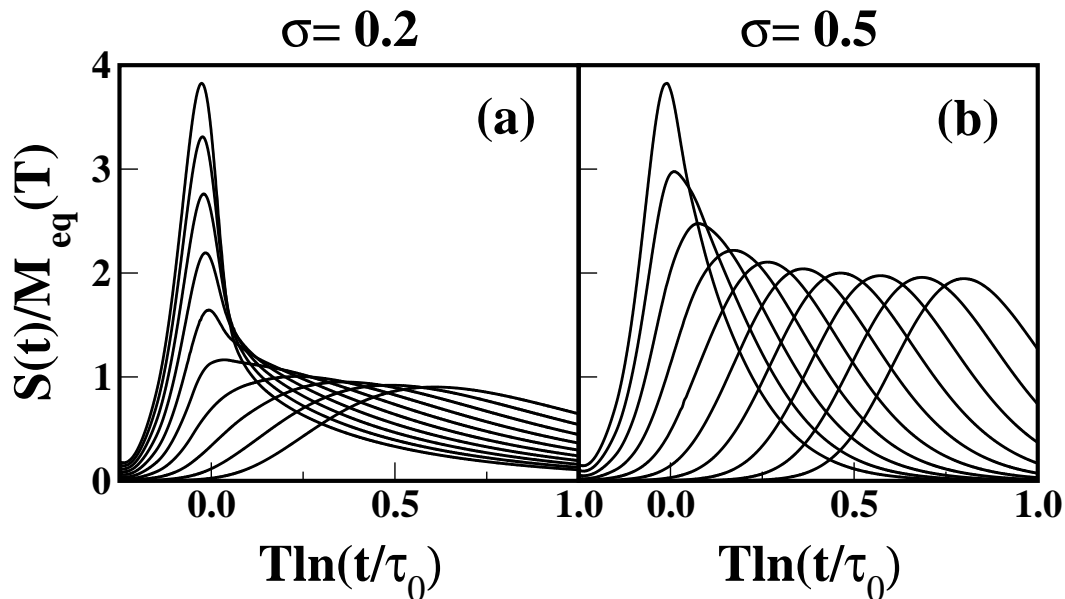


Fig. 4. Low temperature ( $T = 0.05$ ) viscosity curves for a system of particles with random orientations and lognormal distribution of anisotropies with (a)  $\sigma = 0.2$  and (b)  $\sigma = 0.5$ . The curves have been normalized to the equilibrium magnetization and correspond to magnetic fields  $H = 0.1$  to  $1.0$  in  $0.1$  steps starting from the right.

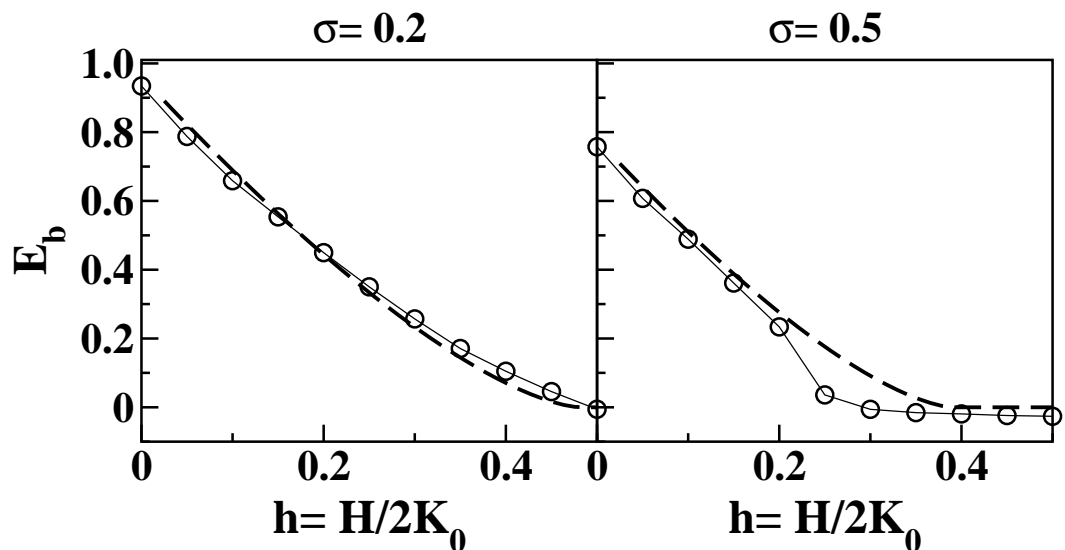


Fig. 5. Field dependence of the energy corresponding to the maximum relaxation rate,  $T \ln(t_{\max}/\tau_0)$ , as derived from the viscosity curves in Fig. 4 for  $T = 0.05$  (circles). The dashed line shows the  $h$  dependence of the lowest energy barriers for the particles oriented at  $\psi = \pi/4$  and  $K$  equal to the most probable value.

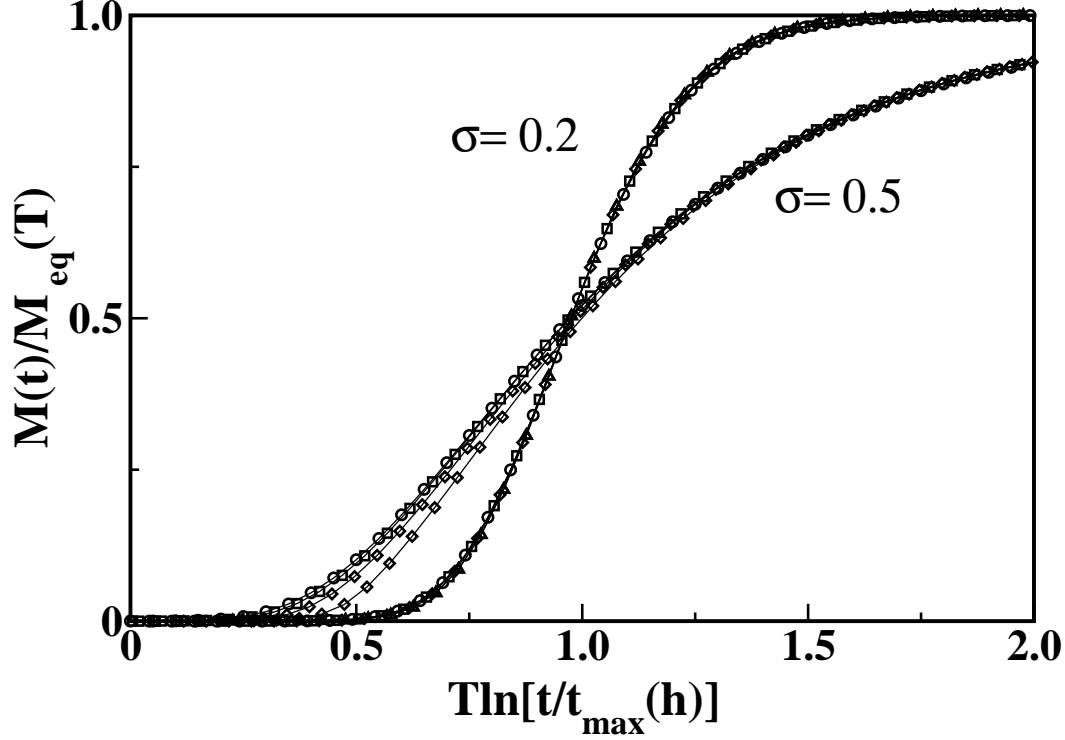


Fig. 6. Normalized relaxation curves as a function of the scaling variable  $T \ln[(t/t_{\max}(h))]$  for  $T = 0.05$ ) obtained from Fig. 4 by shifting the curves in the horizontal axis with the position of the maximum relaxation rate of Fig. 5. For  $\sigma = 0.2$  the field are  $H = 0.1, 0.2, 0.3, 0.4, 0.5$ , and for  $\sigma = 0.5$ ,  $H = 0.1, 0.2, 0.3, 0.4$  (starting from the uppermost curve).

for  $\sigma = 0.2, 0.5$ . They are the equivalent of the master curves of Fig. 3 for a fixed  $h$  and different  $T$ . Now the appropriate scaling variable is

$$E_{\text{sca}} = T \ln[t/t_{\max}(h)] , \quad (11)$$

which generalizes the scaling at fixed  $T$  (another approach to a similar scaling law may be found in [22]). This new scaling is valid for fields lower than  $h_0$ , the field at which the lowest barriers start to be destroyed and above which the relaxation becomes dominated by almost zero energy barriers. Thus, as already discussed in the previous paragraphs, the wider  $\sigma$ , the smaller the  $h$  range for the validity of field scaling.

## 5 Conclusions

We have proposed a model for the relaxation of small particles systems under a magnetic field which can be solved numerically from analytical expressions and which allows to study the effect of the magnetic field on the energy barrier

distribution. In particular, we have shown that the original energy barrier distribution  $f(E_b)$  is split into two subdistributions which evolve towards higher and lower energy values, respectively, as  $h$  increases.

For fields smaller than the critical values for the smallest barriers, the relaxation curves at different  $h$  and fixed  $T$  can be collapsed into a single curve, in a similar way than  $T \ln(t/\tau_0)$  scaling for curves at fixed  $h$ . Whereas the latter allows to extract the barrier distribution by differentiation of the master curve [14], the shifts in the  $T \ln(t/\tau_0)$  axis necessary to produce field scaling, give the field dependence of the mean relaxing barriers, a microscopic information which cannot easily be inferred from other methods [23].

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